Gold in Solar Cells

A REVIEW OF CURRENT WORK

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The cost of producing electricity from solar energy by means of solar cells is still high and limits the use of such devices to special situations. Gold has important roles not only in older types of cells but also in newer types which are being developed to achieve increased efficiencies.

Solar cells are solid state generators that provide current by direct conversion of sunlight into electrical energy. These devices literally rose to fame when the first satellites were launched and space research experiments were powered by them. As we enter the 1980's, solar cells are still playing a key role in powering satellites and the few trials with alternatives are unlikely to be repeated after the experience in 1978 of the problems associated with the uncontrolled re-entry above Canada of a nuclear-powered satellite.

During the space race of the 1960's, the use of gold for contacts and connections in solar cells was the norm because price was no object and because gold has such high electrical conductivity and resistance to corrosion. The latter property means that gold contacts and connectors are reliable in use; it also makes it easy to bond gold to itself and other metals in electronic devices (1). Now that solar cells have come down to earth and the race is on to develop cheaper and cheaper products, gold contacts are becoming less competitive for use in silicon single crystal *p-n* junction devices.

The target of the United States Department of Energy is to produce solar cell modules at a cost of U.S. \$0.70 per watt by 1986 (2). If this goal is to be achieved with the current technology using single crystal cells, in which near-perfect arrangement of the atoms in the crystals and high purity are necessary, the price of the contact material and the package must be kept as low as possible, because the crystal-growing processes that have been developed are relatively expensive. However, using the new, cheaper materials and processes that are emerging, gold, gold-plated or gold-clad contacts could become more common again (3).

There are solar cells, other than those based on silicon single crystals, in which gold has an important place. In the Schottky barrier, gold plays a key role as a part of the heart of the device. In concentrator cells which operate at high temperature, gold contacts can be used with single crystal *p-n* junction cells such as

those made from gallium arsenide (GaAs). Also, experimental cells with new materials often use gold contacts because their properties are so reproducible.

The *p-n* Junction Solar Cells

Before discussing the mode of operation of *p-n* junction and other solar cells, it is appropriate to define some of the terms that will be used in this connection.

The Fermi level of a solid is the energy level below which all electron states are full of electrons, and in *n*-type material to which electron-donating impurities — for instance, phosphorus, arsenic or antimony in silicon — are added in controlled quantities, the Fermi level is close to, or within, the conduction band. In the conduction band, electrons are free to move, but in the valence or bonding band they are bound together with the atoms of the crystal and only the holes can move. Holes are positive charges which occur where electrons are missing. They are produced when impurities which deplete the valence band of electrons — for instance boron, aluminium, gallium or indium in silicon — are added to the crystal. E_{VB} is the energy at the top of the valence band and E_{CB} is the energy at the bottom of the conduction band. The energy gap E_g of a semiconductor crystal is the energy difference between E_{VB} and E_{CB} . Electron- or hole-current flow does not occur in this gap and so it is sometimes referred to as the forbidden gap.

The value of E_g determines to a large extent the properties of the semiconductor and for sunlight absorption the optimum is 1.5 eV, which corresponds to a wavelength of light of 827 nm. Silicon, which to date has been the most commonly used material in *p-n* junction cells because it was developed so far in advance of any other semiconductor material, has an energy gap of 1.1 eV and is not the ideal material for highly efficient solar devices.

A *p-n* junction cell is illustrated in Figure 1 before and after incidence of sunlight. In (a) the cell is in the dark. There can be no net current flow because the Fermi level shown by the dotted line has the same

energy on both sides of the junction. Φ_B is the height of the potential barrier at the junction between the n - and p -type materials. In Figure 1(b), light of energy equal to or greater than E_g shines on the crystal and generates electron-hole pairs at the junction: electrons are excited out of the valence band into the conduction band and leave holes behind. These electrons can now travel down the barrier, as shown by the arrow, and drop into empty states in the conduction band. Some of these photogenerated electrons are shown as negative charges in the conduction band. Similarly, the photogenerated holes move up the barrier and are shown by the additional positive charges. In this non-equilibrium situation, with the light shining on the crystal, the Fermi level on the n side is separated from the Fermi level on the p side by qV , where q is the electron charge and V is the generated photovoltage. Since the quasi Fermi level on the n -side is close to the conduction band, the potential barrier at the junction between the n - and p -type materials is now reduced to $\Phi_B - qV$. The photocurrent flows from the p side (positive charge- or hole-dominated) to the n side (negative charge- or electron-dominated). Thus, a current is generated which flows in the opposite direction to that flowing through the junction when light is excluded by packaging or encapsulation and the device is used as an ordinary semiconductor diode with the positive terminal of the power supply connected to the p side.

In order to collect the photocurrent, two electrodes which make ohmic contact to both the front and back of the cell are required. Ohmic contacts are junctions which are not sensitive to light and which in this case must also be low in resistance. The back electrode is the less critical and is usually made relatively thick in order to keep the resistance low. The front electrode which contacts the n -type semiconductor region, has to collect photogenerated carriers efficiently and transmit sunlight to such an extent that cell efficiency is not reduced appreciably. To achieve this, the front electrode must either consist of a conductive film which is thin enough over the central area of the cell to be transparent to light, and thick enough at the edges to facilitate current collection, or it must consist of a thick conductive mesh bonded to the cell surface. In the latter case, the mesh may be created by a photo-engraving process. The calculation of the optimum mesh size is not simple. Electrical resistance, light transmission and cell area must be optimized. Normally, compromise values for the thicknesses of the electrode and the mesh gaps are established from theory. Irrespective of the design of front electrode used, gold is the conductive material of choice. It is reproducible from a resistivity and fabrication point of view, and solar cells having a very high yield can be produced using it. Moreover, an added advantage of

Fig. 1 Generation of a photovoltage V at a p - n junction in a semiconductor.

(a) At equilibrium in the dark, the Fermi level is constant through the junction.

(b) In sunlight, electron-hole pairs are generated by photons at, or near, the junction and the Fermi levels in this non-equilibrium condition are displaced by qV , where q is the electron charge and V is the generated photo-voltage

gold is that it is corrosion-resistant and can stand the high temperatures to which some cells are subjected.

In addition, the front surface of the cell is finished with an anti-reflection coating. This is a very thin, optically transparent film that 'traps' the sunlight by reducing reflection losses and thereby increases the efficiency of the cell. The thickness of this film has to be accurately controlled and must be very uniform. If it is pinhole-free, it also protects from environmental pollution that portion of the front surface of the cell which is exposed by the mesh spaces.

Fig. 2 Comparison of the load characteristics of a $\text{Cu}_2\text{S}/\text{CdS}$ p - n junction solar cell in sunlight with the 'dark' current-voltage curve of the cell under forward bias, that is when the p side of the junction is connected to the positive of a battery. V_{OC} is the open circuit photovoltage and I_{SC} is the short circuit photocurrent

Fig. 3 Calculation of the efficiency of a $\text{Cu}_2\text{S}/\text{CdS}$ p - n junction solar cell from its load characteristics

Figure 2 compares the normal current-voltage (I-V) relationship of a p - n junction (its dark characteristics) with that when it operates in the solar mode (its light characteristics). The light characteristics show the I-V relationship below the open circuit voltage, V_{OC} , which is the voltage developed across the cell when there is no load resistor across it. The characteristics for voltages between 0 and V_{OC} are measured after the introduction of a variable load resistor across the cell. For this reason, curves such as the bottom one of

Fig. 4 Variation of the theoretical efficiency with the energy gap E_g of the material used for p - n junction solar cells

Figure 2 are sometimes referred to as the load characteristics to avoid confusing them with the full I-V characteristics, which are obtained by applying both positive and negative bias by means of a battery or power supply placed across the junction. Discussion here is restricted to the load characteristics.

The short circuit current, I_{SC} , is the current with the cell shorted out with zero load resistance. I_{SC} is directly proportional to the light intensity, whereas V_{OC} saturates when this reaches a 'one sun' value, that is $100 \text{ mW}/\text{cm}^2$. For all p - n junction solar cells, I_{SC} depends on both the area of the cell and the light intensity, and saturates at very high light intensities. In contrast, V_{OC} increases very rapidly at low intensities and then begins to level out at an intensity of about one third of the 'one sun' value.

The calculation of the efficiency of a solar cell from its load characteristics is shown in Figure 3. The maximum power point P, at which the cell works most efficiently, is determined by drawing the tangent APB to the curve such that $AP = PB$. (This can be proved simply by theory (4)). The maximum power in watts is then PC multiplied by PD . In Figure 3, this is equal to 0.323 W . As the cell area is $7.4 \times 7.4 \text{ cm}$, the power per unit area is $5.9 \text{ mW}/\text{cm}^2$. Since $100 \text{ mW}/\text{cm}^2$ is falling on the cell, its efficiency is 5.9 per cent.

Silicon p - n junction solar cells with efficiencies well in excess of 10 per cent are now made, but silicon will never be the best material for high efficiency cells because of its low energy gap. Figure 4 shows a theoretical curve of p - n junction solar cell efficiency against energy gap value (5, 6). Silicon is compared to GaAs and CdTe, and it can be seen that these latter compounds, which have an energy gap close to the

theoretical optimum, should make higher efficiency cells. This has been borne out by some initial experiments which have shown that GaAs *p-n* junction cells with efficiencies of over 20 per cent can easily be made (7). In the case of silicon, efficiencies up to only 18 per cent have been achieved (8), after more than twenty years of intensive research.

In concluding this schematic description of the *p-n* junction cell, it is worth mentioning two of its shortcomings. First, as is obvious from what has been said, its output power is proportional to the sunlight intensity, so that in countries in which this varies, considerably less solar power will be obtained and it will be difficult to run solar systems without battery backup during the day. Secondly, the operating temperature of cells of this type is important because it affects their efficiency (Figure 5).

Cu₂S/CdS Cells with Gold Contacts

For over twenty years, gold has been used in contacts to polycrystalline copper sulphide/cadmium sulphide (Cu₂S/CdS) *p-n* junction cells. It is used for the front contact in the manner illustrated in Figure 6, which shows light falling on a low-cost flexible cell (9).

To prepare this type of device, a CdS film is deposited onto a conductive substrate of metallized polyester by electrophoresis from a sol obtained by bubbling hydrogen sulphide into a solution of cadmium acetate in water. Water is used in preference to alcohol as medium because it has a high dielectric constant and aqueous sols are therefore more stable than sols in an alcohol. A CdS particle size of about 10 nm is aimed for, to inhibit flocculation which would preclude the formation of pinhole-free thin films. The water sol is diluted with an alcohol just before electrophoresis to prevent excessive oxygen and hydrogen emission at the electrodes. Anodic deposition onto a polyester electrode metallized with stainless steel is the most common procedure, but gold and platinum metallizations have also been tested. However, in the case of platinum, very poor adhesion caused film cracking during drying when the CdS film was baked in air to harden it after deposition. CdS films about 1.5 μm thick are usual because flexible, pinhole-free deposits of this thickness can be prepared. The flexibility is required to allow use of cheap plastic substrates.

The Cu₂S film is then evaporated onto the CdS and must not be thicker than 200 nm or the light will not penetrate to the junction region where the photocurrent is generated. In order to complete the circuit, front and back contacts must be established with the Cu₂S and CdS films respectively. The back contact is provided by the metal film between the CdS and the polyester support. The front contact must be shaped so that it screens off a minimum of the Cu₂S film

Fig. 5 Variation of the efficiency of a Cu₂S/CdS *p-n* junction solar cell with temperature

Fig. 6 Schematic section of a Cu₂S/CdS *p-n* junction solar cell with a gold-plated copper mesh top contact. The CdS layer is deposited electrophoretically

from incident sunlight and its 'window geometry' must therefore be carefully designed. In practice, either a gold-plated copper mesh is bonded to the Cu₂S with a gold-containing conductive epoxy adhesive or a pressure contact plate is applied onto the front surface. The gold coating on the copper is deposited either by electroplating or by ion plating (10). Screen printing of gold mesh contacts has also been carried out successfully.

The colour photograph in Figure 7 shows experimental coatings on 25 μm thick polyester films. Various designs of solar cells of this type have been studied, using both gold and other metals as the back contact materials. Efficiencies have always been low, however, and much more work will be required if the dream of a low-cost flexible solar cell that can be mass-produced by a continuous process and stored easily is to materialize. Moreover, gold electrodes on

Fig. 7 Thin films deposited on polyester for research on flexible solar cells. From right to left the films are CdS on steel, gold, copper and aluminium. In every case, except for the CdS film which was deposited electrophoretically by the method described in the text to a thickness of $1.5\ \mu\text{m}$, the films are $200\ \text{nm}$ thick

CdS were never found to be ohmic but were found to form effective Schottky barriers.

The Gold Schottky Barrier Solar Cell

The solar cells discussed so far make use of the photovoltaic effect across the interface between a p - and an n -type semiconductor. Schottky barrier solar cells rely upon a similar effect across the interface between a metal and a semiconductor.

Fig. 8 Schematic cross-section of a gold Schottky barrier solar cell

The cross-section of one type of gold Schottky barrier solar cell is shown in Figure 8. The device consists of a 5 to $20\ \text{nm}$ thick gold film which is evaporated or ion-plated onto the freshly etched surface of an n -type semiconductor. A thicker gold contact dot is then evaporated onto the gold electrode and the sequence is completed by an anti-reflection oxide coating to increase the absorption of light, and thereby enhance the efficiency of the cell. A gold wire, attached to the contact dot by thermocompression bonding, collects the current from the front or gold side of the cell and the circuit is closed by a connection to the back or semiconductor side of the cell.

The thickness and structure of the gold film are important, since its resistivity must be as low as possible and it must be transparent to solar light. The significance in this latter regard of the deposition process and conditions is clearly shown by the set of curves in Figure 9, in which the resistivity of gold films on glass is plotted as a function of thickness and of the bias voltages used in ionized-cluster beam evaporation technology (11). Clearly, films formed with a $4\ \text{kV}$ bias consisted of close packed gold atoms and gave the lowest resistance.

The energy band diagram shown in Figure 10 indicates how a current flow is generated in a gold Schottky barrier exposed to light. For simplicity, the

Fermi level in the gold is shown equal to the Fermi level in the semiconductor at the instant that the light falls on the cell. The barrier height is Φ_B before the incidence of sunlight. After the electron-hole pairs are photogenerated, this height is reduced to $\Phi_B - qV$. The Fermi levels are then separated by qV as in Figure 1. The band bending and the depletion region where it occurs can be understood by considering what happens at the metal/semiconductor interface. Electrons are depleted from the semiconductor in the region of the junction, so the Fermi level must move away from the conduction band. The bands therefore bend to accommodate for this change in concentration.

In the case of gold/silicon Schottky barrier cells, it has been shown that the gold migrates into the semiconductor. Gold is electrically active in silicon and changes its conductivity, it is therefore a highly undesirable impurity. To overcome this problem, a thin oxide film is grown or deposited onto the silicon to prevent the diffusion of gold. The resultant gold/oxide/silicon devices operate very well and work is in progress on them in the United States. These are strictly speaking metal/insulator/semiconductor (MIS) cells, but their theory is very similar to that of Schottky barriers.

The I-V characteristics of the Schottky barrier solar cell at constant light intensity are very similar to those of the simple p - n junction. However, Schottky barrier cells do have some disadvantages when compared with the latter (12):

- (1) As surface devices, their performance is critically dependent on the formation of very thin pinhole-free oxide and barrier-metal layers
- (2) Their theoretical efficiency, even when there is an interfacial insulating layer (MIS-type devices), is lower
- (3) The lifetime of cells that have been produced so far is not very good. There is a tendency for the load characteristics to drift with time, unless extreme precautions are taken during barrier formation to exclude contaminants like water vapour
- (4) Their surface is fragile, which means that relatively expensive evaporation techniques only can be used for applying top contacts. Cheaper screen printed gold contacts cannot be used since they have to be cured thermally and since heating of the cell after the gold/semiconductor barrier has been formed causes degradation.

Nevertheless, the following advantages over silicon single crystal p - n junction cells justify continued research on Schottky barrier solar cells (12):

- (1) Lower temperature processes are required for their manufacture, so less energy is used in preparing the cells
- (2) This lower processing temperature means that cheaper polycrystalline silicon can be used

Fig. 9 Variation of the resistivity of gold films on glass as a function of their thickness and the bias voltage used for depositing them by cluster ion beam techniques. The resistivity of bulk gold is shown for comparison purposes

Fig. 10 Photovoltaic effects at a gold Schottky barrier

because little diffusion of impurities along the grain boundaries occurs

- (3) Thin film preparation techniques such as ion plating (10) can be used to deposit in a single operation both the silicon and the gold layers on a low-cost substrate material. This process has the potential of being very economical since it can be scaled up with a moving belt and moving electrode system
- (4) There are some materials which cannot be made both *p*- and *n*-type, so Schottky barrier cells are the only solar devices that can be made from them.

Gold in Experimental Solar Cells

After reviewing in general terms the *p-n* junction and the gold Schottky barrier cells, it is now time to discuss some specific examples of how gold is being used in a wide range of experimental solar modules. Since the use of gold is so extensive, the author has limited this section to describing the work presented at the Conference held in Luxembourg in September, 1977 (13). A recent Solar Energy Research Institute Conference held in Denver (14) confirmed that most of this research work with gold is continuing and even expanding.

Kipperman *et al.* (15) reported on a novel gold/silicon Schottky barrier cell formed on *p*-type silicon. It was of the MIS-type and the novelty lay in the way the silicon oxide was formed. Instead of using the conventional dry oxidation technique, these authors used a new hydrofluoric acid wet treatment to produce a pinhole-free insulating layer between the semiconductor and the gold. The oxidizing liquid was prepared by dissolving SiO_2 in a 20 per cent hydrofluoric acid solution for about two minutes. *p*-type silicon of conductivity 2 ohmcm was used and the back contact was a titanium-gold alloy electrode. The thin transparent gold front electrode was DC-sputtered in argon at a pressure of 5.3 Pa. Finally, a smaller thick gold contact was added, so that strong electrical contacts could be made to the front gold layer. The thin gold electrode had a resistance in the range 14 to 20 ohms per square. Open circuit voltages in excess of 300 mV were measured for the optimum oxide thickness and structure, but no efficiency figures were given. A trapping-assisted tunnelling model (16) was used to explain theoretically the behaviour of these cells.

The Institute of Energy Conversion at the University of Delaware has always used gold top contacts in work on $\text{Cu}_2\text{S}/\text{CdS}$ *p-n* junction solar cells. It is the largest group working on solar devices and still believes that evaporated CdS cells will be the first truly low-cost devices on the market. At Luxembourg, members of this group reported on an improved gold top contact technology (17). To obtain a good

contact with the Cu_2S they evaporated a gold grid and then pressure-bonded on top of this in an orthogonal arrangement another gold grid prepared by electrodeposition. The optical transmission of the hybrid was 91 per cent only, but the high current collection efficiency thus achieved helped to raise the overall efficiency of the cells close to 8 per cent and good reproducibility was obtained. The Delaware group has now reached close to 9 per cent efficiency using fully evaporated gold grids with 95 per cent optical transmission and silicon monoxide anti-reflection coatings. Shorting-out problems with the current collection tab contact have been overcome by depositing an insulating layer under the tab.

The Delaware group used thick (20 to 40 μm) evaporated CdS coatings on zinc-plated copper substrates acting as back contacts, but a German group (18) used for this purpose an evaporated silver film on a glass substrate. As top contact, a gold-plated copper foil grid was bonded onto the Cu_2S and a cover glass was then sealed down onto the grid in such a way that total encapsulation was achieved. No anti-reflection film was used and for large 7 x 7 cm cells, efficiencies of 4 per cent were achieved. It was also reported that the gold-plated grids were very stable under conditions where nickel-plated grids showed considerable degradation.

Gold is a very popular contact material for GaAs solar cells, because it appears to produce no contamination of the semiconductor. For example, in a *p-n* junction cell with a *p*-type GaAlAs window, contacts have been made to the *n*-type GaAs base of the cell by means of evaporated gold-germanium alloy (19). With a titanium dioxide anti-reflection coating, an efficiency of 15.9 per cent was achieved under 'one sun' illumination.

GaAlAs/GaAs cells for use in concentrated sunlight up to the 600 sun level and using gold in both the bottom and front electrodes were also described (20). Evaporated indium-germanium-gold alloys were used for contacts to the *n*-type base and titanium-gold alloys for contacts to the *p*-type front window. A grid pattern was photo-engraved onto the titanium-gold and the grid thickness was then increased to 3 μm by gold electroplating. Anti-reflection coatings were produced by anodizing the GaAlAs window. The solar cell chips were mounted on standard transistor headers and thermocompression-bonded gold wires were used to make contact to the header pins. Gold plating of the Kovar base gave low electrical and thermal resistance values.

Cadmium telluride (CdTe) which has an energy gap close to the optimum for solar cells (see Figure 4), has received an increasing amount of attention recently. Gold contacts to *p*-type CdTe in CdTe/CdS solar cells have been applied by two methods (21):

- (1) Evaporation after etching of CdTe in sulphuric chromic acid solution. The gold film is then annealed in hydrogen at about 250°C
- (2) Replacement from a gold chloride solution.

The highest efficiency achieved with CdTe/CdS cells was 7.9 per cent. In addition to CdTe/CdS cells, CdTe/Cu₂Te cells have been made and, in this latter case, gold contacts were again deposited either by evaporation or from a gold chloride solution.

Gold Schottky barriers have been formed on evaporated polycrystalline films of cadmium selenide (CdSe) about 3 µm thick (22). A 10 nm thick insulating layer which consisted of an oxide, a fluoride or a high energy gap semiconductor like zinc sulphide was deposited on the CdSe before the transparent gold films were finally evaporated. The thickness of the gold contacts was not optimized, but it ranged from 10 to 30 nm giving an optical transmission of 50 to 20 per cent. Gold was also used as the base contact in these experimental cells.

Two new semiconductor materials, tungsten selenide (WSe₂) (23) and zinc phosphide (Zn₃P₂) (24) have also been used for solar cells. WSe₂ has two absorption edges, at 1.35 and 1.60 eV, and thus has an energy gap very close to the theoretical optimum. It also has very good chemical stability. Gold Schottky barriers have been formed on *n*-type single crystals of WSe₂ without an intentional oxide layer. A 5 nm thick gold film was used to give good optical transparency and an additional 100 nm of gold was evaporated where a contact pad with the Schottky gold layer was required. The back contact was a gallium-indium alloy and an efficiency of just over 1 per cent was achieved without an anti-reflection coating. It is likely that WSe₂ solar cells will be studied more intensely in the future.

Work on *p*-type Zn₃P₂, which is at a very early stage, indicates that it has an energy gap of 1.4 eV, very close to the optimum for solar cells. Gold reacts with Zn₃P₂ and can only be used in MIS-type Schottky barrier configuration for top contacts. Iron appears to be the best material for the base contacts and good results have been obtained using it.

Conclusions

Gold is playing a key role in solar cell research. It is being used for contacts in a variety of devices and as an active constituent of Schottky barrier cells. Such cells are the only kind that can be used in the case of many materials which do not display both *p*- and *n*-type conductivity.

In GaAs concentrator cells, gold is a reliable contact material which withstands the high operating temperatures resulting from concentration of sunlight on the cells. Under such circumstances, the high cost of gold is not an important consideration because the

area required is small in comparison with that of cells which do not use concentrated sunlight.

The use of gold in *p-n* junction cells of large area, like the Cu₂S/CdS thin film cells, will be restricted to a very thin gold coating, say 10 nm thick, deposited on the copper grid or on the Cu₂S surface immediately before the copper grid is applied. As far as silicon solar cells are concerned, gold can be used in Schottky barriers only when an insulating layer has been grown or deposited onto the silicon surface because of changes in electrical properties which are brought about by gold atoms which diffuse into the silicon crystal lattice.

The adoption of new plating procedures in which gold can be applied with very little waste will be essential if the metal is to be retained for commercial solar cells as they become progressively cheaper in the years to come. But in solar cells for space exploration, gold will probably have no competitor as the ideal contact and interconnection material because it is so reliable, so chemically inert and can tolerate the high temperatures on satellite solar power 'arms'.

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